M.R.P. 1012
S.C.111/221
31st October 1956.

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An estimate of the value and practicability of routine measurements

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(Meteorological Office)

METEOROLOGICAL
OFFICE
22 FEB 1957
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METEOROLOGICAL RESEARCH COMMITTEE

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1. Introduction

At the 35th meeting of the Physical Sub-Committee of the Meteorological Research Committee, held on December 8th 1955, it was recommended that the value of regular ground level counts of freezing nuclei at selected stations should be considered. It was decided, therefore, to write a brief review of the position and, in view of an earlier recommendation of the Sub-Committee (29th meeting, April 2nd 1954), to consider also condensation nuclei and dust particles.

2. Dust particles

2.1. Dust is most usually collected for examination by directing a jet of dust-containing air onto a glass plate. Sometimes, as in the Owens counter, the air is first moistened to assist adhesion, but this seems not to be necessary if the speed of air through the jet is high. The deposit can afterwards be examined in various ways. If it is not too heavy, it can be examined under a microscope and the particles counted: but it is evident that the result of the count depends mainly on the resolving power of the microscope and has, therefore, little meteorological or physical significance.

2.2. F. Teichert (11) has very recently published a detailed study of methods of sampling and measuring dust, and he dismisses counting as a useless procedure. He shows by photographs how every increase in resolution of the microscope shows a very large proportional increase in the number of particles. To obtain a significant result, he says "one would have to observe the number of particles of each size and shape, so that the counting of a single dust-sample would be a whole day's work".

2.3. Even a simple count, it appears, is a fairly difficult task. Unfortunately, it has not been possible to consult anyone who has personal experience, but Teichert, after remarking that any apparatus, to be satisfactory, must be manageable by "average meteorological personnel" adds "counting of the particles, therefore, is out of the question".

2.4. It is certainly true that, although counts of dust particles have been made for many years, they have not been a very productive field of meteorological study. They have some relevance to problems of visibility, though even here the mere number of particles is not very useful unless the size-distribution is known, but they are of very little interest in the study of condensation and precipitation.

2.5. If it were decided to make observations of this character, the Owens dust counter would be the most readily available instrument. It is simple and robust, and the cost of equipment would not be great - probably less than £100 per station. But in view of the small meteorological interest of the results and the practical difficulties of counting, it is doubtful whether the attempt would be justified.

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3. Condensation nuclei

The condensation nuclei of direct interest in meteorology are those which are responsible for the formation of cloud and fog. Since, so far as is known, relative humidities appreciably exceeding 100 per cent never occur in the atmosphere, the only nuclei of significance in meteorology are those which act at, or not much above, saturation.

3.1. The nuclei most commonly studied, however, have been the Aitken nuclei - nuclei activated by an instantaneous expansion resulting in supersaturations of anything from 10 per cent to 300 per cent according to the type of instrument. These Aitken nuclei are far more numerous than the true condensation nuclei, their concentrations ranging from a few thousand to a few hundred thousand per cc, whereas there are usually only one or two hundreds of cloud droplets per cc. So far, there is no evidence of any useful relationship between the number of Aitken nuclei and the number of the true condensation nuclei.

3.2. H. Dessens remarked (1) of the Aitken nuclei - "The majority ..... are instrumental nuclei, defined, and perhaps created, by a particular type of apparatus: their influence on clouds and precipitation remains hypothetical".

3.3. This is a somewhat harsh judgment: the nuclei are certainly not created by the instrument, since filtered air gives no nuclei. Moreover, provided a supersaturation exceeding 10 per cent is employed, the count is almost independent of the supersaturation until very high values are reached. It can fairly be inferred that expansion instruments of the Aitken type do count everything that could be described either as a droplet or a solid particle and that they measure a real property of the air. Nevertheless, Dessens' final sentence is certainly justified, and the study of Aitken nuclei, though of some interest in itself, seems unlikely to contribute very greatly to other branches of meteorology. A glance at the titles of the papers read at the first Symposium on Condensation Nuclei (3) held in Dublin in 1955 immediately reveals the isolation of the subject. Moreover, a study which is primarily concerned with the number of nuclei, only indirectly with their size-distribution, and hardly at all with their constitution, seems a very incomplete one. One may foresee that it will eventually be linked closely with the rapidly developing subject of atmospheric chemistry, and that results of much greater scientific interest will emerge when the constitution of the particles is studied in relation to their size and number.

3.4. It might, nevertheless, be worth while to make regular measurements of Aitken nuclei at a few points in order to obtain some preliminary information on their dependence on synoptic conditions or other meteorological parameters. Similar work is being carried out at several places in Europe, and there should be opportunities for interesting comparisons. Such studies, however, entail a great deal of laborious statistical work and it is unlikely that any progress would be made unless at least one officer could devote a substantial proportion of his time to it.

3.5. The original Aitken counter, and others which resemble it in principle and method of observation, are very difficult to use, and not suited to routine observations. Pollak (2) has developed a photoelectric nucleus counter which is easy to manipulate and which can readily be made completely automatic. In this instrument a beam of light passes down a long tubular chamber, the walls of which are kept moist, and falls on a photocell at the far end. A sample of air is admitted, and a quantity of filtered air is pumped in to produce an over pressure. When sufficient time has elapsed for the air to become saturated, the over pressure is released and a cloud forms, the extinction coefficient of which is measured by the photocell. The concentration is then determined by an empirical calibration based on a comparison with an Aitken-type instrument.

3.6. All the operations of this measurement can be carried out by automatic valves and switches, and the results recorded on, for example, a recording milliammeter. Components would cost something under £100, and the apparatus could

be assembled in the workshops at Kew Observatory, only about two weeks' work being required per instrument.

3.7. Hourly observations would be convenient for most purposes: for fog studies or other special purposes it would be easy to increase the rate to six per hour.

3.8. Sites which are not very liable to pollution from local fires, etc. are desirable. Lerwick, Eskdalemuir, Aberporth and St. Eval suggest themselves: the first two are not quite free from local pollution in the direction of the prevailing wind, but on the other hand they have Scientific Officer staff, which may be advantageous since one can hardly expect a newly-designed apparatus to give no trouble.

3.9. It might be thought more realistic to study, not the Aitken nuclei, but those which are active at rates of cooling comparable with those occurring in nature. The number of nuclei activated, however, increases rapidly as the rate of cooling is increased, so that the properties of the air-sample could only be represented by a table or curve relating these two quantities. This is not impossible, but the experiment is necessarily long and complicated, and would be unsuitable for performance as routine.

4. Freezing nuclei

It is generally accepted that freezing nuclei play an important part in the formation of precipitation, and it has been widely assumed that their presence or absence exercises an important degree of control over rainfall. Measurements of their availability must therefore be of great interest and importance; it is only necessary to decide (a) what quantity should be measured and (b) how the measurement should be made.

4.1. In considering a sample of air containing freezing nuclei, we require to know the number of nuclei which will induce freezing at any given temperature, as the sample is progressively cooled. Superficially, this appears to be a simple problem; it should only be necessary to take a sample of air, induce a cloud of droplets to form in it, and then cool it: the measurement would consist in noting from time to time the temperature which had been reached and the number of ice-crystals which had been formed.

4.2. Many such experiments have been carried out, and the results of some of them, including the best-known ones, are shown in Fig. 1 (reproduced from (4)). The enormous discrepancies, which are obvious at a glance, are certainly not real for the most part, but represent faults in the instruments. These have never been very fully or satisfactorily analysed: but the following are believed to be among the principal reasons for the lack of agreement.

4.2.1. The principal source of discrepancies, and the one of greatest theoretical interest, is concerned with the manner in which the droplet-cloud is formed. A freezing nucleus can only act if it is within a droplet - in other words, before it can act as a freezing nucleus, it must already have acted as a condensation nucleus.

4.2.2. If, then, the cloud is formed by gradually cooling the air-sample, either by external refrigeration or by expansion, droplets will form only on the particles which are active (as condensation nuclei) at low values of supersaturation. The result will be a cloud consisting of a comparatively small number of large droplets, formed on a particular class of particle. If a greater rate of cooling is employed, a higher degree of supersaturation will be momentarily produced, condensation will take place on more particles and therefore more will be able, if they have the right crystalline structure, to act as freezing nuclei.

/4.2.3.

4.2.3. The importance of the effect has, apparently, never been studied in detail. It is evident that a high supersaturation should lead to a greater total number of active nuclei, but its effect on the form of the freezing-nucleus spectrum - (that is the curve relating the number of crystals present to temperature) cannot be judged a priori. Findeisen and Schulz (5) used two different rates of expansion, equivalent to rates of ascent in the free atmosphere of 5 m/sec and 20 m/sec, and obtained two completely different spectra (Fig.2). In particular, ice crystals appeared at much higher temperatures when a low rate of expansion was used.

4.2.4. The tendency observed by Findeisen and Schulz is confirmed by the observations of Cwilong (6), who used a Wilson type of cloud chamber and almost instantaneous cooling: and found that no ice crystals appeared at temperatures higher than -27 to -32°C.

4.2.5. It appears from this discussion that the nucleating properties of a sample of air could only be satisfactorily described by a set of freezing-nucleus spectra, each one corresponding to a different rate of expansion - the relevant rate of expansion being of course that occurring at the moment when the cloud is formed.

4.2.6. A second possible source of discrepancy is the phenomenon of regeneration. Palmer (7) and others have demonstrated that ice crystals, once formed, have a certain power of regeneration by splintering. Findeisen and Schulz (5) noticed a phenomenon which may well be related to this -

"In many experiments, in which only moderately low temperatures are reached and therefore the number of ice crystals is in general still small, there appear occasional swarms of ice-particles, about 10-20 fragments, which are concentrated in a relatively small volume".

4.2.7. This suggests a possibility that the number of ice-crystals present may exceed considerably the number of nuclei present before freezing begins. It is evident that the longer we keep a growing ice-crystal under observation the more likely we are to see one of these explosions, if indeed they occur, and it may not be only chance that Findeisen and Schulz, who used an unusually deep chamber, are apparently the only experimenters to observe the phenomenon. Two things follow:-

(a) if one wishes to know the number of freezing nuclei present, one should use a fairly shallow cloud-chamber so as to reduce the risk of regeneration;

(b) the number of ice-crystals observed in a sample of air in a cold chamber may bear very little relationship to the number which would appear in the same air under natural conditions, even at the same temperature and with the same rate of cooling. On the whole the phenomenon seems unlikely to produce major discrepancies between the results of different instruments, although it may create serious difficulties in the application of the results to natural conditions.

4.2.8. Bigg (8) has shown that, at least in the case of pure water, there is not a definite temperature at which a given drop will freeze. There is only (in the case of a continuously cooled drop) a certain probability that the drop will freeze at any selected temperature below 0°C, and this probability depends upon the rate of cooling. It is probable, though so far as is known it has not been demonstrated, that the same rule holds for droplets containing freezing nuclei. If so, the form of the freezing-nucleus spectrum will be considerably affected by the rate of cooling of the sample at all stages of the experiment.

4.3. The difficulties which have been mentioned so far are fundamental to the problem - they would arise even if one could observe the behaviour of freezing nuclei in a natural cloud. In addition there are many instrumental problems.

4.3.1. Perhaps the most important is the settling both of droplets and of ice-crystals. If one operates with a chamber which is gradually cooled a great many drops, and especially the larger ones, settle-out and are lost before they crystallise. Again crystals are constantly settling-out, and if the quantity observed is, as usually, the number of crystals in suspension, this is the result of a balance between the rate of formation and the rate of settling, and there is no satisfactory way of relating this to the total number which has been formed. E.K. Bigg (private communication) has recently found a satisfactory

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means of overcoming this difficulty. He allows the crystals to fall to the bottom of the chamber, where they are caught in a shallow layer of a concentrated solution of sugar in water at a temperature of about -12°C. The crystals grow rapidly in this solution, each one developing within a few minutes into a perfect hexagon several millimetres across: these can be very easily photographed or counted. The method, evidently, is satisfactory only at low crystal-concentrations - one per cc at most - but these are in fact the concentrations of greatest interest. The problem of the settling-out of ice-crystals can, therefore, be regarded as solved: the settling-out of water droplets remains.

4.3.2. Even more important is the condensation of water on the walls of the chamber. This is bound to occur in any chamber where cooling is produced by cooling the walls, and takes place on a very large scale. Indeed, all such cloud chambers have to be constantly supplied with additional water, in the form of steam, in order to maintain the cloud - and this is in addition to the water released by cooling. This condensation must remove very large numbers of freezing nuclei from the air. Indeed, H. Cauer (9) has used the deposition of moisture on a polished metal surface as a particularly efficient method of collecting condensation nuclei.

4.3.3. When the cloud chamber is used for the study of freezing nuclei, periods of twenty minutes or more may elapse between the formation of the fog and some of the measurements. In this time the mass of water deposited will usually be comparable with, and may even exceed, the whole mass originally present, and the result of the experiment may be profoundly modified as a result of the loss of condensation nuclei. (Findeisen and Schulz appear not to have recognised this danger, perhaps because they believed that they were dealing with sublimation nuclei: at all events, in some experiments they deliberately dried the air by passing it through narrow tubes immersed in a cooling fluid.)

4.3.4. There are usually very large temperature differences between different parts of the experimental chamber - differences quite comparable with the whole range of temperature under investigation - and it is difficult to know what temperature to assign to a particular observation of crystal-concentration. Usually both temperature and concentration are measured near the centre of the chamber: but the air is in constant movement, and it is quite likely that the crystals one sees in the centre were formed close to the walls, where the temperature was several degrees lower.

4.4. It may be useful now to consider, in the light of what has so far been said, what sort of observation would be necessary and adequate for routine use.

4.4.1. The observation would need to yield a complete nucleus spectrum for a specified rate of expansion - a single constant rate of expansion throughout the experiment would probably be adequate.

4.4.2. Such an observation would be sufficient only if means can be found to adjust the nucleus spectrum for other rates of expansion.

4.4.3. The cloud chamber would need to be free from large internal temperature differences and designed so as to eliminate or correct for settling-out, deposition on the walls, and regeneration.

4.4.4. The means of counting crystals should be objective.

4.5. Without going into details, one may say at once that none of the methods of which accounts have yet been published approaches this specification. In particular, the only equipment of which the Meteorological Office has any experience, the "cold box" described in Ref. (1), has every one of the defects described in paras. 4.2. and 4.3. and it is impossible to feel much confidence in the significance of the variations registered by it.

/4.6.

4.6. Since the complete problem appears so intractable, it is natural to seek some simpler and more easily measured, but still useful, quantity. Thus Schaefer (10) has for some years made regular counts on Mount Washington of the nucleus concentration at a particular temperature (-18°C). The quantity thus measured is not a very useful one, being merely one point on the freezing-nucleus spectrum, the whole of which is needed for any useful application to the physics of precipitation. The Mount Washington count can be shown to vary from day to day, but the significance of the variations cannot be estimated.

4.7. Much more promising is the threshold temperature originally proposed by Cwilong (5). This is simply the highest temperature at which freezing nuclei are active. Many freezing-nucleus spectra show an increase in concentration from zero to several hundred per litre occurring within a few degrees Centigrade: and it is evident that if this were the general rule the threshold temperature would be the only important parameter; one would be sure that the whole of a natural cloud above the level of the threshold temperature was completely glaciated, and that all precipitation elements (or at least all those due to the Bergeron-Findeisen process) were derived from this level.

4.8. The value of the threshold temperature is unfortunately suspect for two reasons.

4.8.1. There is reason to think that it may vary considerably with the rate of cooling used to form the cloud. Some evidence on this question has already been presented in para. 4.2.1.- 4.2.4.

4.8.2. It is not certain that a considerable concentration of ice-crystals does in general develop immediately below the threshold temperature. In the observations made by the Meteorological Research Flight in January 1956 (4), for example, it was generally found that a few crystals, about 20/m^3, were observed at relatively high temperatures, -5°C to -12°C. The number fluctuated irregularly but showed no systematic increase until a temperature of about -20°C was reached, when there was a rapid increase to a concentration of about 1 per litre or more. A third sharp increase, to a value of 10^4 - 10^5 per litre, occurred at about -26°C. It is not suggested that the results of the M.R.F. are any more reliable than any of the others presented in Fig. 1: they have been examined in detail only because they happened to be readily accessible. Nevertheless, if taken at their face value, they suggest that the concentration increases in a slow and irregular (though by no means unsystematic) manner, and that there is in many cases no definite and clear-cut threshold temperature. The value of threshold temperature measured is, in these circumstances, likely to depend on the volume of air examined. If the instrument holds, say, only 200 cc of air, concentration of less than one particle per litre will generally be missed. This means that, taking the M.R.F.'s measurements as reliable, the observed threshold temperature is likely to be around -26°F and that the crystals formed at higher temperatures will be unnoticed. Yet these crystals may well be highly important, for in fact clouds are generally completely glaciated at temperatures of -16°C and often at considerably higher temperatures.

4.9. But in spite of these doubts, the threshold temperature is evidently deserving of further study, if only because of the great difficulty of finding any other manageable parameter. The principal objection, stated in para. 4.8.1, would vanish if it could be shown that there were a law connecting the threshold temperature with supersaturation.

4.10. Very recently E.K. Bigg (unpublished) has introduced a most interesting type of cloud chamber. It consists of a cylindrical vessel of ten litres capacity, maintained by external refrigeration at a temperature of -11°C. The bottom is entirely covered by a shallow dish containing sugar solution. The air to be sampled is admitted to the vessel, suitably compressed, and allowed to take up the temperature of the vessel. The pressure is then released,

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causing adiabatic cooling to some predetermined temperature, and a cloud, containing some ice-crystals, results. The crystals fall into the sugar solution. After a few minutes the cover is removed from the instrument and the crystals, now grown to an easily-visible size, are counted.

4.10.1. This instrument fulfils an unusually high proportion of the conditions set out in para. 4.4. above. It yields a complete nucleus spectrum for a given rate of expansion (i.e., for instantaneous expansion). Internal temperature differences are small because no observation windows are necessary, and therefore the chamber can be completely surrounded by the coolant. No error arises from settling-out of crystals; nor even from the deposition of moisture provided the air is originally dry enough to be cooled to -12°C without condensation occurring (it will therefore be most reliable when used in an aircraft or on a mountain where the temperature is low). The chamber is large enough to ensure detection at concentrations of one per litre or even less but small enough to make it probable that regeneration is negligible. In fact, only two doubts - both very serious ones certainly - remain. These are

(a) Can results obtained with instantaneous expansion be corrected in some way to make them applicable to rates of expansion comparable with those occurring in nature?

(b) Are serious errors introduced because the minimum temperature attained as a result of expansion is held for an instant only, as Bigg's researches (8) suggest may be the case?

4.10.2. Evidently some research on these two problems is desirable. Meanwhile, it can only be said that of all the instruments which have been employed hitherto, this appears by far the most promising. Dr. E.G. Bowen (C.S.I.R.O.) has kindly offered to supply an instrument for use in this country, probably early in 1957, and it will then be possible to form a better idea of the instrument's capabilities.

5. Conclusions

5.1. Counts of dust particles by means of the Owens dust counter appear to have no significant applications to general meteorology, and it is doubtful whether they would justify the expense and time which they would demand.

5.2. Counts of Aitken nuclei likewise seem to have no significant applications unless supplemented by information on their size and nature. The subject is however developing rapidly and is receiving a great deal of attention in many counties. It is moreover possible that interesting relationships with other meteorological parameters will reveal themselves. Provided, therefore, that sufficient time can be allocated to the search for and study of such relationships, it is considered that it would be reasonable to begin regular counts at one or two places which are relatively free from man-made pollution. The counting can be done automatically and takes up little time.

5.3. Counts of condensation nuclei active at rates of cooling comparable with those occurring in the free air would probably be of greater interest than counts of Aitken nuclei, but the problem would necessarily be extremely complex and has not yet been attacked at all systematically.

5.4. Counts of freezing nuclei would undoubtedly be of very great interest, if they could be made under natural conditions. This however presents an extremely difficult practical problem, largely because the count is a function of so many variables.

/5.5.

5.5. The most readily measurable quantities appear to be

(a) the threshold temperature

(b) the complete nucleus spectrum for instantaneous adiabatic cooling.

Neither of these quantities can be applied with confidence to problems of precipitation, but it is possible that further research may reveal laws which will enable them to be so applied, or interesting relationships with other meteorological parameters.

5.6. No satisfactory instrument for measuring threshold temperature as routine has yet been designed - indeed there is not even a clear and accepted definition of threshold temperature - but the problem does not appear intractable.

5.7. Bigg's apparatus for measuring the nucleus spectrum for instantaneous cooling is not entirely unsuited to routine use, but is rather slow to use (about 15 minutes for each point on the curve) and moreover would need skilful operators.

5.8. In view of these difficulties, it is suggested that

(a) the Bigg cloud chamber which has been promised us by C.S.I.R.O. should be installed and operated as routine in some suitable station - again Lerwick or Eskdalemuir seems to be indicated.

(b) an instrument on the same principle should be designed and fitted in the M.R.F. Hastings or Varsity.

5.9. Experiments of the kind just indicated, however, can by themselves amount to little more than groping hopefully in the dark: the significance of their results cannot be assessed unless they are supported by fundamental researches on the behaviour of both condensation and freezing nuclei, particularly on the variations in the number of nuclei with variations in the rate of cooling. Such researches are likely to be difficult and to require somewhat complex apparatus: they would require the full attention of one Scientific Officer for at least two years. In addition, of course, the study of the results of freezing-nucleus measurements, which would necessarily consist of statistical comparisons with other parameters, would require a great deal of time.

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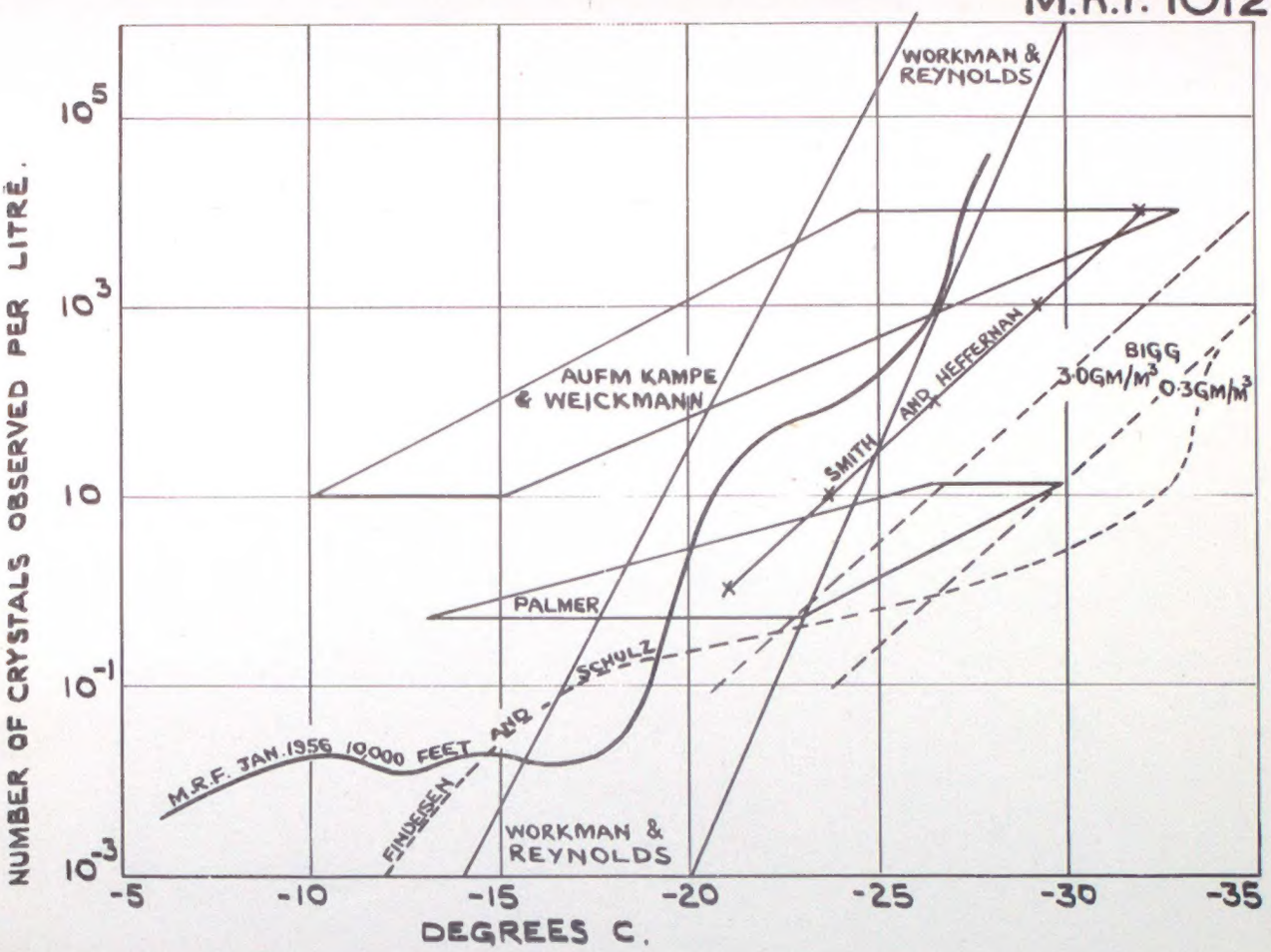

FIG.1. Concentration of freezing nuclei in air. Comparison of various measurements.

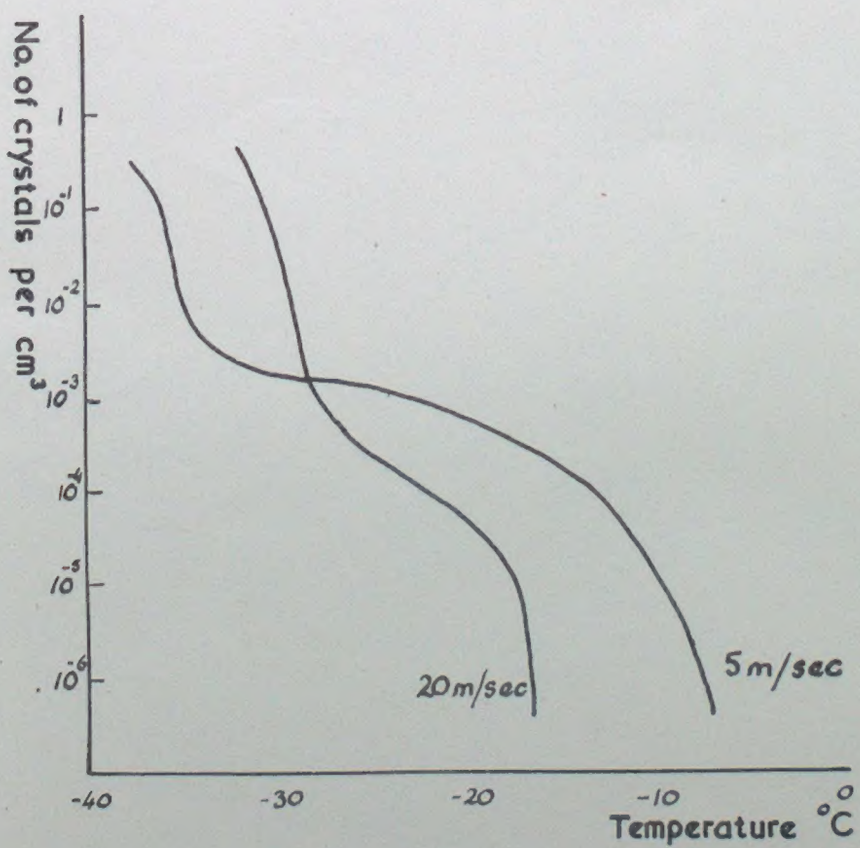

FIG.2. Average ice nucleus spectrum for surface air at Prague, obtained with rates of expansion corresponding to vertical speeds of 5m/sec and 20 m/sec. (after Findeisen and Schulz).